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# Observed Aerosol Effects on Marine Cloud Nucleation and Supersaturation

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**Abstract.** Aerosol particles in the marine boundary layer include primary organic and salt particles from sea spray and combustion-derived particles from ships and coastal cities. These particle types serve as nuclei for marine cloud droplet activation, although the particles that activate depend on the particle size and composition as well as the supersaturation that results from cloud updraft velocities. The Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE) 2011 was a targeted aircraft campaign to assess how different particle types nucleate cloud droplets. As part of E-PEACE 2011, we studied the role of marine particles as cloud droplet nuclei and used emitted particle sources to separate particle-induced feedbacks from dynamical variability. The emitted particle sources included shipboard smoke-generated particles with 0.05-1 μm diameters (which produced tracks measured by satellite and had drop composition characteristic of organic smoke) and combustion particles from container ships with 0.05-0.2 μm diameters (which were measured in a variety of conditions with droplets containing both organic and sulfate components) [1]. Three central aspects of the collaborative E-PEACE results are: (1) the size and chemical composition of the emitted smoke particles compared to ship-track-forming cargo ship emissions as well as background marine particles, with particular attention to the role of organic particles, (2) the characteristics of cloud track formation for smoke and cargo ships, as well as the role of multi-layered low clouds, and (3) the implications of these findings for quantifying aerosol indirect effects. For comparison with the E-PEACE results, the preliminary results of the Stratocumulus Observations of Los-Angeles Emissions Derived Aerosol-Droplets (SOLEDAD) 2012 provided evidence of the cloud-nucleating roles of both marine organic particles and coastal urban pollution, with simultaneous measurements of the effective supersaturations of the clouds in the California coastal region.

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# EASTERN PACIFIC EMITTED AEROSOL CLOUD EXPERIMENT (E-PEACE) 2011

E-PEACE combined a targeted aircraft campaign off the coast of Monterey, California, in July and August 2011, with embedded ship (Fig. 1) and satellite observations and modeling studies. Atmospheric conditions in the northeastern Pacific during July are ideal for formation of homogeneous layers of persistent stratocumulus clouds. The layers observed have consistent diurnal characteristics, cloud thicknesses of 100 to 300 m, and cloud top heights typically below 500 m. We employed the R/V *Point Sur* to measure the aerosol below cloud and as a platform for well-characterized smoke emissions to produce a uniquely identifiable cloud signature. The Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft was used with a full payload of instruments to measure particle and cloud number, mass, and composition.

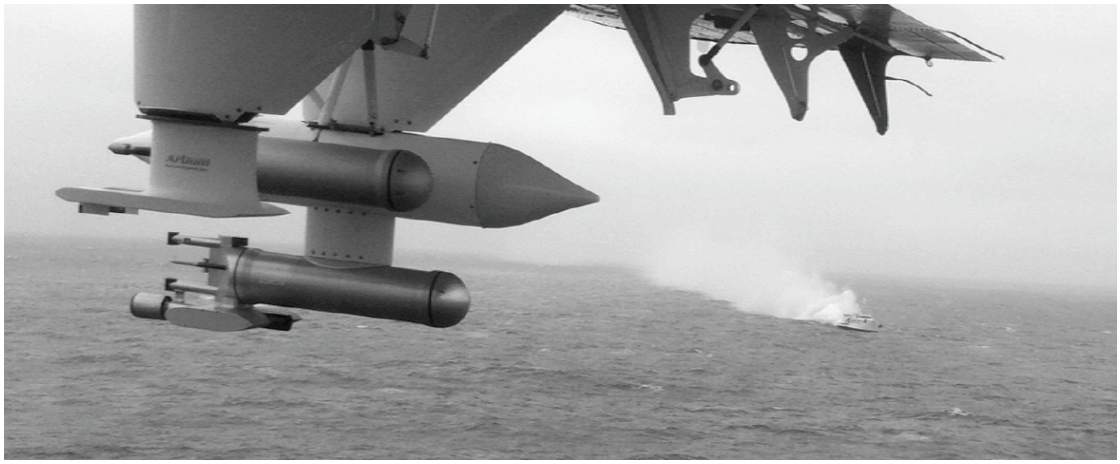

**FIGURE 1.** R/V *Point Sur* from the CIRPAS Twin Otter during E-PEACE 2011, showing the persistence of the plume of smoke generated on the ship in the atmosphere and some of the aircraft instruments for measuring particles and clouds (Photograph modified from Russell et al. [1]).

The Twin Otter aircraft flew into clouds behind the emissions from ships. Figure 2 shows one example of measurements of the number of particles below cloud and droplets in cloud, and the pie graphs show that these droplets were almost entirely organic components with trace amounts of sulfate. The measured ship and marine characteristics of the organic components during E-PEACE were used to quantify the widespread contributions of ship emissions to the marine boundary layer aerosol [2]. The counterflow virtual impactor (CVI) was used as an inlet for evaporating droplets as they were brought into the aircraft, allowing sampling of droplet chemical composition [3]. The large organic fraction in Fig. 2A is characteristic of smoke emitted from the generators on the R/V *Point Sur* and contrasts with the composition

of droplets in the cloud not affected by the smoke (Fig. 2C), which are made up of three-quarters sulfate and few organic components.

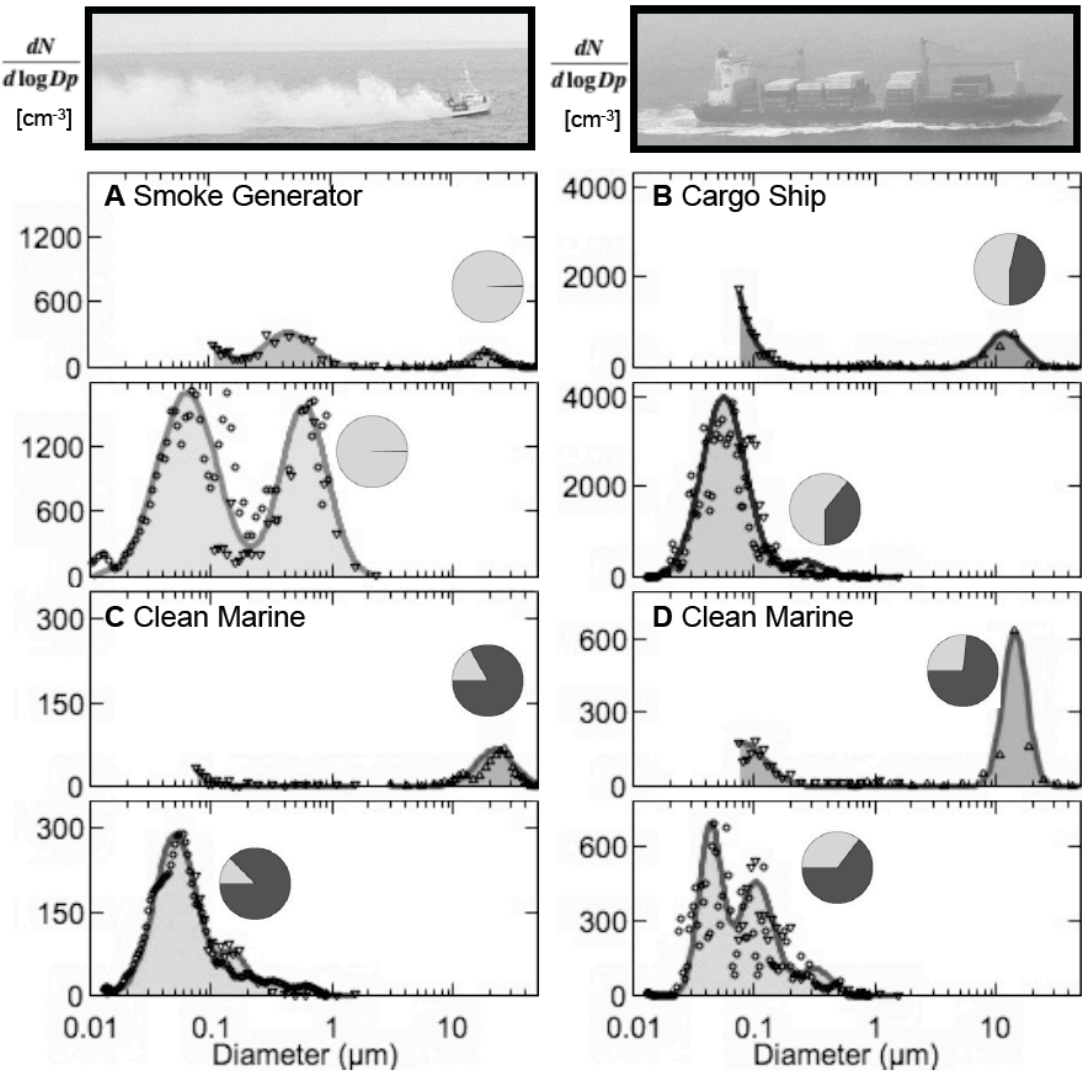


**FIGURE 2.** Examples of particle and droplet number distributions and mass-based non-refractory chemical composition, from measurements below (bottom panel) and in (top panel) cloud, for the smoke generator on the R/V *Point Sur* on 16 July (panel A) and for the stack emissions of a cargo ship on 10 August (panel B). The background particle and droplet concentrations are shown for 16 July and 10 August (panels C and D, respectively). The size distributions are plotted at the measured relative humidity, wet for supermicron droplets in cloud (triangles-up: 3 μm < CDP < 50 μm for 16 July and 1 μm < CAS < 50 μm for 10 August), with passive heating for submicron particles in (interstitial) and below cloud (triangles-down: 0.1 μm < PCASP < 2 μm), and dried below cloud (diamonds: 0.01 μm < Scanning DMA < 0.9 μm). The pies show composition of the droplets in cloud measured by AMS for submicron particles below cloud (bottom panel) and for the residuals of cloud droplets (top panel) that are left after drying in a counterflow virtual impactor (11 μm < CVI), with light grey for organic components and dark grey for sulfate). Refractory chemical components (such as Sea Salt) were not measured behind the CVI and are not included in the pie graphs. The measurements were collected on the CIRPAS Twin Otter on 10 August for the cargo ship (1651-1831) and 16 July for the smoke generators (1704-1801). (Reproduced from Russell et al. [2], used with permission. ©American Meteorological Society)

It is interesting that the background cloud droplets also have 10-30% organic components. Since the organic composition *outside of* the smoke and ship plumes was very similar to characteristic organic functional groups associated with marine sources [4], it is likely that this organic fraction was of marine origin. The presence of a similar organic fraction in the droplet residuals shows that this organic mass also acted as CCN in nucleating cloud drops.

To understand the differences in the two days shown in Fig. 2, we compared the measured updraft velocity and estimated the maximum supersaturation by comparing the cloud drop number (CDN) concentration with the cloud condensation nuclei (CCN) spectra. The results for the two days are given in Table 1, which is taken from Russell et al. [1]. While there is uncertainty in using the maximum supersaturation calculated from the measured average CDN and the CCN spectra, the calculated updraft velocities were consistent with the measured maximum updraft velocities (in cloud) of 0.94 m $s^{-1}$ (respectively) on 16 July and 1.2 m $s^{-1}$ (respectively) on 10 August.

**TABLE 1.** Particle and droplet characteristics for below and in-cloud measurements shown in Fig. 2.

| | **Measured Maximum Updraft Velocity [m $s^{-1}$]** | **Calculated Maximum Supersaturation [%]** |
|---|---|---|
| 16 July 2011 (generator smoke) | 0.94 | 0.09 |
| 10 August 2011 (cargo ship) | 1.22 | 0.25 |

## STRATOCUMULUS OBSERVATIONS OF LOS-ANGELES EMISSIONS DERIVED AEROSOL-DROPLETS (SOLEDAD) 2012

Stratocumulus Observations of Los-Angeles Emissions Derived Aerosol-Droplets (SOLEDAD) 2012 took place during May and June in La Jolla, California. Aerosol particles were sampled at Scripps pier and at the peak of Mount Soledad, 1.5 mi from the pier sampling location and 250 m above sea level. Stratocumulus clouds were observed frequently at levels between 100 and 500 m, and there were several nighttime cloud layers during which the mountaintop site was in cloud. One such event is shown in Fig. 3. These events tended to be characterized by onshore winds and CDN concentrations below 100 $cm^{-3}$, suggesting that the clouds were similar to the marine stratocumulus sampled during E-PEACE 2011.

The SOLEDAD 2012 campaign provided the opportunity to measure drop formation in coastal conditions and to calculate supersaturation by a second method. In this campaign, several hours of sampling in cloud allowed us to use measurements of CCN concentration behind the counterflow virtual impactor to compare the measured supersaturation of the droplet residuals to the CDN concentration. With this approach, we measured a distribution of supersaturations during up to 6 hr of sampling in each cloud event. The values ranged from below 0.1% to above 0.7%, although most were below 0.4%. It is possible that the larger supersaturations reflect the contributions of scavenged components to the droplet composition, but it is also likely that this range reflects the variability in updraft velocities.

**FIGURE 3.** Instrumented van on Mount Soledad during a stratocumulus cloud measured as part of SOLEDAD 2012. The picture shows the rotating aerosol inlet, the counterflow virtual impactor, in-cloud spectrometers, and an active-flow cloud water collector.

## CLOUD NUCLEATION AND SUPERSATURATION

For both campaigns, online measurements of non-refractory components in aerosol particles below cloud and droplet residual in cloud show very similar compositions – in other words the similarity of droplet and submicron aerosol composition in multiple marine stratocumulus cloud events suggests that that the accumulation mode mass is responsible for most of the nucleating droplets. While some droplet compositions will be changed by scavenging of interstitial particles, the very similar fractions of different components suggests that the nucleated particles are largely the same as the submicron particles in composition. A striking example is provided by the drop residuals composed of generated smoke nuclei, since their composition indicates the ability of very "fresh" hydrocarbon emissions to nucleate cloud droplets at low supersaturations with negligible hygroscopic uptake by sulfate.

These results were found for clouds with a range of supersaturations typical of marine stratocumulus, from below 0.1% up to 0.4%. We estimated these effective supersaturations in two ways, both by directly comparing the activated droplet population to the measured CCN spectra and by measuring the CCN spectra of the

droplet residuals. Interestingly, this variability is comparable to the values used to calculate that in-cloud sulfate formation could contribute to new CCN [5].

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